Predicting Mineral Transformations in Supercritical CO_2 Containing Systems: The System Mg^{2+}-H^+-HCO_3^--CO_2-H_2O.

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The capture and storage of carbon dioxide and other greenhouse gases in deep geologic formations represents one of the most promising options for mitigating the impacts of greenhouse gases on global warming. As a result there has been a focus on evaluating mineral-fluid interaction for subsurface CO_2 storage in aqueous solutions in contact with supercritical CO_2 ($scCO_2$). One of the key factors in evaluating mineral-fluid interactions is the potential for the formation of divalent metal carbonates, principally Ca and Mg phases, which can sequester the disposed CO_2 as mineral precipitates. Unfortunately, the formation of some of these phases, especially the anhydrous phases such as magnesite ($MgCO_3$), has been hindered by their well-known sluggish precipitation kinetics owing to the strong hydration energy of the aqueous Mg^{2+} ion. The issue of understanding the precipitation of magnesite is one that may prove pivotal in the development of efficient carbon sequestration techniques. We have recently become the first to discover the formation of magnesite at low temperatures (as low as 35^0C). Hence it is of considerable interest to understand the unique conditions that promote such a transformation and the underlying molecular level mechanisms. Here we present recently obtained experimental data on the formation of magnesite from aqueous solutions saturated with $scCO_2$. These studies were conducted over a range of temperature, pressure, and initial $Mg(HCO_3)_2$ concentration to map out the specific solution phase conditions which result in nucleation of magnesite in aqueous solution at low temperature. The aqueous solution measurements on speciation are currently being utilized to test and enhance the thermodynamic model (MSE) being developed in collaboration with OLI Systems Inc. The measurements made on the $scCO_2$ solutions as part of this study will expand the range of model parameterization beyond the present data set which is mostly limited to subcritical CO_2 gas phase equilibria. Complementary measurements of water solubilities in $scCO_2$ as a function of aqueous electrolyte concentration will also be presented that substantiate the applicability of the MSE model to the CO_2 rich side of the phase diagram.